LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-PROC-210833

Strain Relaxation in Si_(1-x)Ge_(x) Thin Films on Si(100) Substrates: Modeling and Comparisons with Experiments

K. Kolluri, L. A. Zepeda-Ruiz, C. S. Murthy, D. Maroudas

March 25, 2005

Materials Research Society
San Francisco, CA, United States
March 28, 2005 through April 1, 2005

Strain Relaxation in $Si_{1-x}Ge_x$ Thin Films on Si (100) Substrates: Modeling and Comparisons with Experiments

Kedarnath Kolluri[1], Luis A. Zepeda-Ruiz[2], Cheruvu S. Murthy[3], and Dimitrios Maroudas[1]
[1]Department of Chemical Engineering, University of Massachusetts, Amherst, MA 01003, U.S.A.
[2]Chemistry & Material Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, CA 94550, U.S.A.
[3]IBM Semiconductor Research & Development Center, Hopewell Junction, NY 12533,U.S.A.

ABSTRACT

Strained semiconductor thin films grown epitaxially on semiconductor substrates of different composition, such as $Si_{1-x}Ge_x$/Si, are becoming increasingly important in modern microelectronic technologies. In this paper, we report a hierarchical computational approach for analysis of dislocation formation, glide motion, multiplication, and annihilation in $Si_{1-x}Ge_x$ epitaxial thin films on Si substrates. Specifically, a condition is developed for determining the critical film thickness with respect to misfit dislocation generation as a function of overall film composition, film compositional grading, and (compliant) substrate thickness. In addition, the kinetics of strain relaxation in the epitaxial film during growth or thermal annealing (including post-implantation annealing) is analyzed using a properly parameterized dislocation mean-field theoretical model, which describes plastic deformation dynamics due to threading dislocation propagation. The theoretical results for $Si_{1-x}Ge_x$ epitaxial thin films grown on Si (100) substrates are compared with experimental measurements and are used to discuss film growth and thermal processing protocols toward optimizing the mechanical response of the epitaxial film.

INTRODUCTION

Strained Si devices on $Si_{1-x}Ge_x$ virtual substrates enhance electron and hole mobility compared to unstrained substrates of the same material [1]. When an alloyed $Si_{1-x}Ge_x$ layer is grown on a Si substrate, biaxial strain develops due to lattice mismatch between the substrate and the grown film. Possible mechanisms of strain relaxation include misfit dislocation generation at the film/substrate interface [2] beyond a critical film thickness, as well as film surface morphological transitions [3]. In practice, large numbers of threading dislocations are nucleated which, after gliding a short distance, become immobilized, resulting in a high dislocation density in the film. Device-quality materials, however, need to have a high degree of strain relaxation, low threading dislocation densities, and smooth surfaces. Recently, it has been reported that He ion implantation and subsequent annealing at temperatures (T) over the range $1023\ \mathrm{K} \leq T \leq 1123\ \mathrm{K}$ can result in thin $Si_{1-x}Ge_x$ layers possessing a high degree of strain relaxation, as well as relatively low densities of threading dislocations [4].

In this paper, we report a hierarchical approach for computational analysis of the mechanical response of $Si_{1-x}Ge_x$ films on Si substrates. We use continuum elasticity and dislocation theory to study the critical thickness of $Si_{1-x}Ge_x$ films as a function of the alloy composition in an $Si_{1-x}Ge_x$/Si(100) heteroepitaxial system. Subsequently, we employ a phenomenological model to examine the kinetics of strain relaxation during thermal annealing that follows a typical ion implantation process used for the post-growth treatment of

heteroepitaxial films. Finally, we examine the role of atomistic simulations in parameterizing consistently the continuum models for critical film thickness calculation and strain relaxation kinetic analysis.

HIERARCHICAL COMPUTATIONAL APPROACH

Our study focuses on theoretical analysis of the deformation mechanics, interfacial stability, strain relaxation kinetics, and surface morphology of strained-layer $Si_{1-x}Ge_x/Si$ heteroepitaxial systems. Toward this end, we have developed a hierarchical approach combining continuum elasticity and dislocation theory with atomistic simulations of structural and compositional relaxation, within a reliable empirical description of interatomic interactions. Special emphasis is placed on the case of heteroepitaxial growth on compliant substrates of finite thickness. The analysis aims at rigorous parameterization of continuum theoretical models for the mechanical response of strained-layer heteroepitaxial systems. A diagrammatic outline of our hierarchical computational approach is given in figure 1.

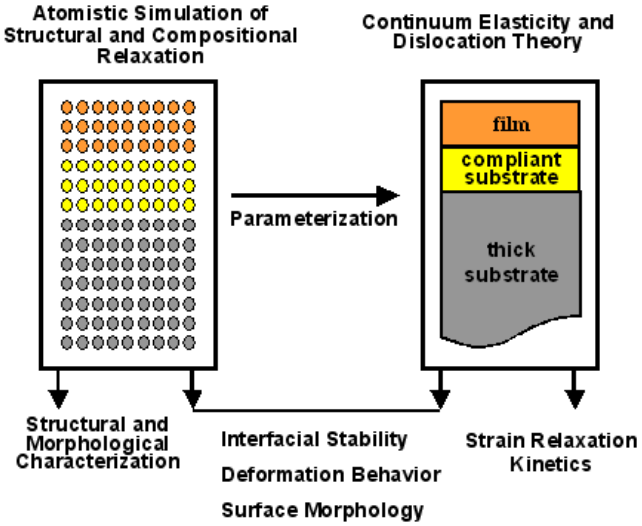

Figure 1. Diagrammatic outline of hierarchical computational approach to study mechanical response of strained-layer heteroepitaxial systems.

CALCULATION OF CRITICAL FILM THICKNESS

First, we consider the case where the epitaxial film of thickness h_f and the substrate of (generally finite) thickness h_s are coherently elastically strained. Taking the equilibrium lattice parameter of the film to be less than that of the substrate, the lattice mismatch results in compressive strain, ε_f, for the film, and tensile strain, ε_s, for the substrate. Assuming uniform deformation, $\varepsilon_{xx,i} = \varepsilon_{yy,i} = \varepsilon_i$ and $\sigma_{xx,i} = \sigma_{yy,i} = M_i\varepsilon_i$, where $i = f, s$, and, M_f and M_s are the corresponding biaxial moduli defined as $M \equiv 2\mu(1+\nu)/(1-\nu)$, where μ and ν are the shear modulus and Poisson's ratio, respectively. The condition of zero net force on any atomic plane perpendicular to the interface requires that $\sigma_f h_f + \sigma_s h_s = 0$, which yields $M_s\varepsilon_s h_s + M_f\varepsilon_f h_f = 0$. In addition, the compatibility condition for perfect interfacial coherence requires $\varepsilon_f - \varepsilon_s = \varepsilon_m$, where ε_m is the mismatch strain in the film in the limit $h_s \to \infty$. Solving the mechanical equilibrium and compatibility conditions for the elastic strain in the absence of dislocations, we obtain $\varepsilon_f = \varepsilon_m/[1+\Lambda(h_f/h_s)]$ and $\varepsilon_s = -\varepsilon_m\Lambda(h_f/h_s)/[1+\Lambda(h_f/h_s)]$, where $\Lambda \equiv M_f/M_s$, is the ratio of the moduli of the film and the substrate.

Next, we consider a dislocation with Burgers vector $\{b_x, b_y, b_z\}$ that is introduced within the substrate directed at the film/substrate interface. The work of the stress done on the dislocation can be expressed as $W_1 = M_s\,\varepsilon_s\,h_s\,b_x$ and the self-energy of dislocation, W_2, according to Ref. [5]. The critical condition for the introduction of a misfit dislocation at the interface requires that $W_1 + W_2 = 0$, which yields

$$\frac{[b_x^2 + b_y^2 + (1-\nu_s)b_z^2]}{8\pi(1+\nu_s)} \frac{(h_s + \Lambda h_{f,c})}{\Lambda h_s h_{f,c}} \ln\left[\frac{4h_s h_{f,c}}{b(h_s + h_{f,c})}\right] - \varepsilon_m = 0 \ , \qquad (1)$$

an equation that can be solved for the critical film thickness, $h_{f,c}$. The work of stress in the film done on the dislocation is $W = M_f \varepsilon_f h_f b_x - W_2$, which can be rewritten as $W = \tau_{eff} h_f b_x$ to define the effective stress, τ_{eff}, on the dislocation. The resulting expression for the effective stress is

$$\tau_{eff} = \frac{M_f \varepsilon_m}{1 + \Lambda(h_f / h_s)} - \frac{M_s[b_x^2 + b_y^2 + (1-\nu_s)b_z^2]}{8\pi(1+\nu_s)b_x h_f} \ln\left[\frac{4h_s h_f}{b(h_s + h_f)}\right]. \qquad (2)$$

The above analysis is a simple extension of the Freund & Nix theory for compliant-substrate heteroepitaxial systems [5], which yields the well-known Matthews-Blakeslee result [6] for infinitely thick substrates. Figure 2 shows the results of the analysis for the dependence on the substrate thickness of the critical thickness of $Si_{1-x}Ge_x$ films on Si substrates for various film compositions, x. The results of figure 2 were obtained by solving the nonlinear algebraic equation for $h_{f,c}$, Eq. (1), numerically for given h_s and x and carrying out a parametric study over a broad range of h_s and x. The beneficial effects of using thin compliant substrates are evident in figure 2, considering the critical film thickness as a typical metric for strain relaxation. To generate the results of figure 2, we have assumed, as a first approximation, that the film properties (lattice parameter and elastic moduli) vary linearly with the Ge composition, analogous to Vegard's law. A more rigorous dependence of the film properties on the Ge composition can be obtained using atomistic simulations (as described below) for coherently strained systems.

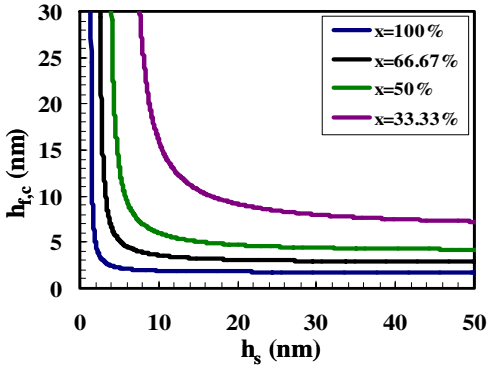

Figure 2. Dependence of the critical film thickness, $h_{f,c}$, on the substrate thickness, hs, and the film composition, x, for heteroepitaxial $Si_{1-x}Ge_x$/Si(100) systems.

MODELING OF STRAIN RELAXATION KINETICS

To study the strain relaxation kinetics after the onset of dislocation generation, we have adopted a variant of the phenomenological model proposed by Alexander and Haasen to describe plastic deformation dynamics in semiconductor crystals [7,8]. In our formulation, the speed of a gliding dislocation, $V(t)$, is given by

$$V(t) = V_o \left[\frac{\tau_{eff}}{\mu_f}\right]^m \exp\left(-\frac{Q_v}{k_B T}\right), \qquad (3)$$

where V_0 is a mobility pre-exponential factor, τ_{eff} is the effective stress, Q_v is the Peierls activation barrier [9], k_B is the Boltzmann's constant, T is temperature, and the exponent $m = 2$ for $Si_{1-x}Ge_x/Si$ [7]. The rate of dislocation generation is given by

$$\frac{dN(t)}{dt} = BN(t)\left[\frac{\tau_{eff}}{\mu_f}\right]^n \exp\left(-\frac{Q_n}{k_B T}\right), \tag{4}$$

where B and n are material constants, N is the dislocation density, and Q_n is the activation barrier for dislocation nucleation. The strain relaxation rate in the film is given by Orowan's equation

$$\frac{d\varepsilon_f(t)}{dt} = N(t)V(t)\cos\lambda \tag{5}$$

The effective stress appearing in Eqs. (3) and (4) is reduced further from the expression of Eq. (2) by subtracting the term $\alpha\mu_f b N^{1/2}$ to take dislocation-dislocation interactions into account; α is a case-dependent numerical constant that can be used as a fitting/adjustable parameter and b is the magnitude of the Burgers vector. In addition, we assume that the deformation remains biaxial as strain relaxation occurs through dislocation formation. The validity of this assumption depends on the surface orientation and is guaranteed for $Si_{1-x}Ge_x/Si(100)$. Finally, in simulating growth experiments we take $dh_f/dt = V_g$, where V_g is the growth velocity and is assumed to be constant.

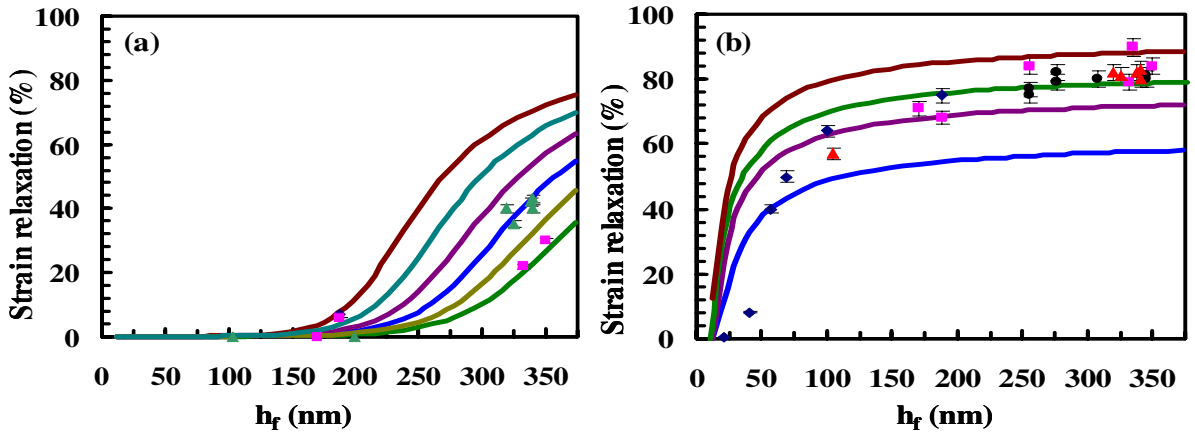

Figure 3. Strain relaxation as a function of film thickness for $Si_{0.80}Ge_{0.20}/Si(100)$ samples annealed after epitaxial growth unimplanted (a) or after He ion implantation (b). The solid curves correspond to the modeling results, while the discrete points correspond to experimental data; different symbols correspond to different experimental conditions [from Ref. 4].

We have integrated Eqs. (3)-(5) to model the thermal annealing experiments reported by Cai, et al [4], where $Si_{0.80}Ge_{0.20}/Si(100)$ samples were annealed after epitaxial growth either unimplanted or following post-growth He ion implantation. For quantitative predictions, we have used dislocation parameters from the literature [7] and taken the experimental conditions carefully into account. During growth, we have used the elastic equations discussed above to model the deformation mechanics for film thicknesses less than critical. Adjusting the parameter α, we maintained a low dislocation density at the end of the growth process ($N < 10^3$ cm^{-2}). For modeling the annealing of unimplanted samples, the initial values of the dislocation density and

film strain were taken equal to the corresponding final values from the kinetic modeling of epitaxial growth. Ion implantation causes substantial dislocation nucleation; therefore, for modeling the annealing of the He implanted samples, we used initial values for the dislocation density higher by orders of magnitude than those obtained from the kinetic modeling of growth.

The modeling results and comparisons with the experimental data are shown in figure 3 for annealing of unimplanted samples, figure 3(a), and annealing after sample implantation with He ions, figure 3(b). The agreement of the modeling results with the experimental data is good and comparable to the agreement reported between the data and state-of-the-art discrete dislocation-dynamics simulations for the case of post-implantation annealing [10].

ATOMIC–SCALE ANALYSIS

Atomic-scale simulations based on accurate many-body interatomic potentials can be used to carry out detailed structural and compositional relaxation and calculate the energy and strain in the relaxed state of epitaxially grown films on thin or thick substrates. These calculations can be used to parameterize phenomenological models of mechanical behavior, which can then be used to evaluate compositional grading schemes in order to grow higher-quality films. The key ingredient of our atomistic simulation procedure is a variant of the Monte Carlo (MC) method originally suggested by Foiles [11,12]. The MC simulation is preceded and followed by energy minimization based on a conjugate-gradient (CG) scheme to account for local structural relaxations. In $Si_{1-x}Ge_x$ systems, the combined MC/CG approach minimizes the system energy by distributing the Si and Ge atoms and relaxing the atomic coordinates after the compositional distribution; therefore, it generates the equilibrium configurations resulting from solute segregation at lattice defects, surfaces, and interfaces. Our MC method employs a three-step sequence: (i) one "compositional" MC sweep over all Ge atoms, where each MC step consists of a trial to exchange a Ge atom with a randomly chosen Si atom (chemical identity switching); (ii) many (typically 50) MC sweeps over all atoms, where each MC step consists of a continuous-space atomic displacement trial for "structural" relaxation; and (iii) one MC step for cell-size ("strain") relaxation, consisting of a trial to adjust the cell dimensions in the principal directions normal to the film's free surface. In all three steps, trials are accepted or rejected according to the Metropolis criterion.

We have implemented the above atomistic simulation method to model the relaxation of a prototypical system, consisting of a $Si_{0.50}Ge_{0.50}$ slab with (100) free surfaces. The interatomic interactions were described according to Tersoff's many-body potential [13]. The results are shown in figure 4 for the evolution of the system energy during relaxation (inset to figure 4(a)), the equilibrated Ge distribution in the slab (figure 4(a)), and the final relaxed atomic configuration (figure 4(b)). The results of figure 4 demonstrate the capabilities of the method to capture the Ge segregation at the slab's surfaces.

SUMMARY AND CONCLUSIONS

We have analyzed the energetics of dislocation formation and the strain relaxation kinetics in $Si_{1-x}Ge_x$ epitaxial films grown layer-by-layer on Si(100) substrates. The critical film thickness for the onset of misfit dislocation formation has been calculated as a function of the substrate thickness and the film composition. A phenomenological models that describes dislocation kinetics during growth and annealing also has been discussed and used successfully

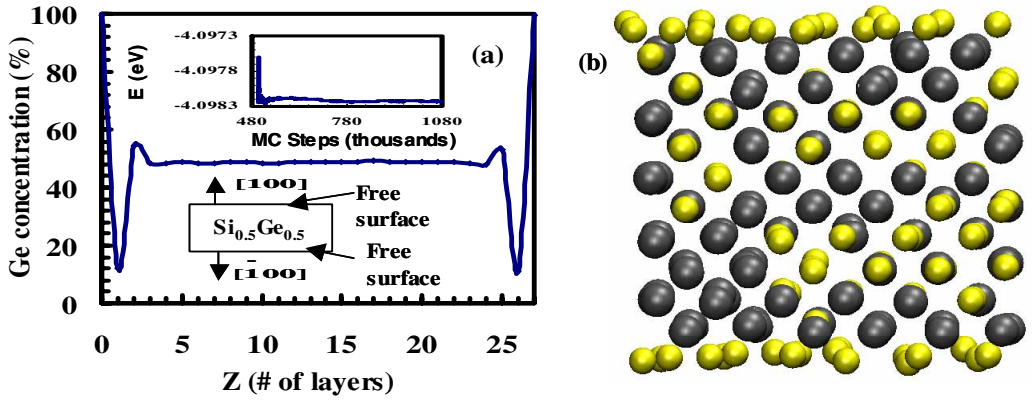

Figure 4. (a) Ge distribution in relaxed configuration of $Si_{0.50}Ge_{0.50}$(100) slab starting from a random distribution of Ge atoms. The inset gives the slab energy evolution during relaxation. (b) Atomic configuration representative of the slab's equilibrated state. Gold and silver spheres denote Ge and Si atoms, respectively.

to interpret experimental data for strain relaxation in $Si_{0.80}Ge_{0.20}$/Si(100) systems. Finally, an atomic-scale method for modeling structural and compositional relaxation in $Si_{1-x}Ge_x$/Si systems toward parameterizing coarse-grained mechanical-behavior models has been presented and demonstrated through a prototypical system consisting of a $Si_{0.50}Ge_{0.50}$ slab with (100) free surfaces; the atomistic simulation has captured the Ge segregation at the slab surfaces.

ACKNOWLEDGEMENTS

Fruitful discussions with K. W. Schwarz and J. O. Chu of IBM's T. J. Watson Research Center, M. R. Gungor, and M. S. Valipa are gratefully acknowledged. This work was supported in part by the National Science Foundation (Award Nos. CMS-0201319 and CMS-0302226).

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

REFERENCES

[1] Y. J. Mii, Y. H. Xie, E. A. Fitzgerald, D. Monrow, F. A. Thiel, B. E. Weir, and L. C. Feldman, *Appl. Phys. Lett*. **59**, 1611 (1991).
[2] J. H. van der Merwe, *J. Appl. Phys*. **34**, 117 (1963).
[3] D. J. Srolovitz, *Acta. Metall*. **37**, 621 (1989).
[4] J. Cai, P. M. Mooney, S. H. Christiansen, H. Chen, J. O. Chu, and J. A. Ott, *J. Appl. Phys*. **95**, 5347 (2004).
[5] L. B. Freund and W. D. Nix, *Appl. Phys. Lett*. **69**, 173 (1996).
[6] J. W. Matthews and A. E. Blakeslee, *J. Crystal Growth* **27**, 118 (1974)
[7] D. C. Houghton, *J. Appl. Phys.* **70**, 2136 (1991).
[8] L. A. Zepeda-Ruiz, B. Z. Nosho, R. I. Pelzel, W. H. Weinberg, and D. Maroudas, *Surf. Sci*. **441**, L911 (1999).
[9] J. P. Hirth and J. Lothe, *Theory of Dislocations* (Wiley, New York, 1982).
[10] K. W. Schwarz, J. Cai, and P. M. Mooney, *Appl. Phys. Lett*. **85**, 2238 (2004).
[11] S. M. Foiles, *Phys Rev. B* **32**, 7685 (1985).
[12] P. C. Kelires and J. Tersoff, *Phys. Rev. Lett*. **63**, 1164 (1989).
[13] J. Tersoff, *Phys. Rev. B* **39**, 5566 (1989).